



I HEREBY CERTIFY THAT THIS CORRESPONDENCE IS BEING DEPOSITED WITH THE UNITED STATES POSTAL SERVICE AS FIRST CLASS MAIL IN AN ENVELOPE ADDRESSED TO: COMMISSIONER FOR PATENTS, P.O. BOX 1450, ALEXANDRIA, VA 22313-1450, ON THE DATE INDICATED BELOW.

BY

*Atsushi Ueda*

Date:

*June 21, 2005*

**MAIL STOP RCE**

**IN THE UNITED STATES PATENT AND TRADEMARK OFFICE**

In Re Patent Application of:	:	
Atsushi Ueda et al.	:	
Conf. No.:	7373	Group Art Unit: 1745
Appln. No.:	10/058,707	Examiner: Raymond Alejandro
Filing Date:	January 28, 2002	Attorney Docket No.: 10059-404US (P27007-01)
Title:	Non-Aqueous Electrolyte Secondary Battery	

**THIRD DECLARATION OF ATSUSHI UEDA UNDER 37 C.F.R. § 1.132**

I, Atsushi Ueda, declare and state as follows:

1. I am a co-inventor of the invention described and claimed in the above-identified patent application. I am the same Atsushi Ueda who executed first and second Declarations under 37 C.F.R. § 1.32, filed in this application on June 15, 2004 and November 8, 2004, respectively.

2. I am familiar with the above-referenced application, and in particular with the final Office Action dated January 21, 2005 (Paper No. 20050115). I am submitting the present Declaration to overcome the obviousness-type double patenting and § 103(a) rejections of claims 1-6 and 8-15 by demonstrating the different effects observed when solvent mixtures containing:

- a cyclic carboxylic acid ester (A);
- a cyclic carbonic acid ester (C);
- optionally a linear carbonic acid ester (D); and

a cyclic carbonic acid ester (B) containing: (i) both vinylene carbonate (VEC) and vinylene carbonate (VC); (ii) only VC; and (iii) only VEC are used in the non-aqueous electrolyte of a secondary battery.

#### The Examiner's Position and the Purpose of the Declaration

3. The Examiner has taken the position that the closest prior art to the invention includes U.S. Patent No. 6,723,473 ("the '473 patent"); U.S. Patent Application Publication No. 2003/0118913 of Takami et al. ("Takami"), WO 01/03228 ("WO '228"), EP 0 796 510 ("EP '510"), U.S. Patent Application Publication No. 2001/0018152 of Kida et al. ("Kida"), U.S. Patent No. 6,090,506 of Inoue et al. ("Inoue"), and U.S. Patent Application Publication No. 2002/0001756 of Hamamoto et al. ("Hamamoto").

4. In the Office Action dated January 21, 2005, the Examiner rejected claims 1-15 under 35 U.S.C. § 103(a) as being obvious over combinations of Takami and WO '228 in view of EP '510, Kida, Inoue and/or Hamamoto. The Examiner argued that all of the claimed elements are taught or suggested by the proposed combinations of cited references, including the inclusion of VEC in a solvent mixture containing a cyclic carboxylic acid ester (A) and a cyclic carbonic acid ester (C).

5. The purpose of this Declaration is to demonstrate that the addition of a combination of VEC *and* VC to a solvent mixture containing components (A) and (C) and optionally (D) results in a dramatic decrease in the amount of gas generated after cycles relative to the amount of gas generated when *only* VEC *or* VC is included in the solvent mixture with components (A), (C), and optionally (D). These results would not be expected based on the prior art, which does not teach or suggest a solvent containing *both* VEC and VC, and such a demonstration will thus overcome the rejections of claims 1-6 and 8-15.

#### Background and Purpose of the Invention

6. The non-aqueous electrolyte secondary batteries according to the present invention were developed to overcome problems known in the art, such as deterioration of charge and discharge characteristics. The batteries according to the invention have excellent charge and discharge characteristics, particularly at low temperature, and show satisfactory

charge and discharge characteristics, even after being exposed to high temperatures over a period of time.

7. According to the presently claimed invention of claims 1-6 and 8-15, non-aqueous electrolyte secondary batteries which exemplify these properties are realized by using a non-aqueous solvent containing: (A) a cyclic carboxylic acid ester; 0.5 to 20 volume % of (B) a cyclic carbonic acid ester having at least one carbon-carbon unsaturated bond and comprising vinylethylene carbonate (VEC) and vinylene carbonate (VC); and (C) a cyclic carbonic acid ester having no carbon-carbon unsaturated bond. In one embodiment, as recited in claims 8 and 9, the electrolyte also contains a linear carbonic acid ester (D). Cyclic carboxylic acid esters (A) are prone to decomposition by hydrofluoric acid, possibly deteriorating the cycle characteristics of the battery. The use of  $\text{LiBF}_4$  also deteriorates cycle characteristics of the battery because the degree of electrolytic dissociation of  $\text{LiBF}_4$  is small or a film formed on the surface of the negative electrode comprising graphite is weak. However, as shown in Tables 10 and 11 of the present application, the use of both salts in combination, particularly in the claimed ratio, improves cycle characteristics of the batteries and decreases the amount of gas generated, and also results in favorable capacity maintenance rates and cycle life. Further, the use of components (C) and VEC in combination with component (A) results in favorable battery characteristics which overcome these disadvantages. The use of VC and VEC in combination is particularly desirable because VEC suppresses the reaction activity of VC at high temperatures to improve storage characteristics of the battery at such temperatures.

8. In order to demonstrate the effect of including both VEC and VC in the non-aqueous solvent on the amount of gas generated after cycles, two series of batteries were prepared using different solvent mixtures containing the following components:

<u>Series A</u>	<u>Series B</u>
components (A), (C) and VC	components (A), (C), (D) and VC
components (A), (C) and VEC	components (A), (C), (D) and VEC
components (A), (C), VC and VEC	components (A), (C), (D), VC and VEC

The amounts of gas generated after cycles for each of these batteries were measured and compared.

### Experimental Procedure

9. Batteries were produced using the procedure described in Example 6 of the above identified application. Non-aqueous electrolytes were prepared by dissolving  $\text{LiBF}_4$  in each of the solvent mixtures shown in Table 1 at a  $\text{LiBF}_4$  concentration of 1 mol/L.

**Table 1**

Battery	cyclic carboxylic acid ester (A)	cyclic carbonic acid ester (B)	cyclic carbonic acid ester (C)	cyclic carbonic acid ester (D)	solute	solute concentration (mol/L)
<b>SERIES A BATTERIES</b>						
1	GBL 77 vol %	VC 2 vol % + VEC 1 vol %	EC 20 vol %	-----	$\text{LiBF}_4$	1
2	GBL 77 vol %	VC 3 vol %	EC 20 vol %	-----	$\text{LiBF}_4$	1
3	GBL 77 vol %	VEC 3 vol %	EC 20 vol %	-----	$\text{LiBF}_4$	1
<b>SERIES B BATTERIES</b>						
1G	GBL 67 vol %	VC 2 vol % + VEC 1 vol %	EC 20 vol %	DEC 10 vol %	$\text{LiBF}_4$	1
2G	GBL 68 vol %	VC 2 vol %	EC 20 vol %	DEC 10 vol %	$\text{LiBF}_4$	1
3G	GBL 69 vol %	VEC 1 vol %	EC 20 vol %	DEC 10 vol %	$\text{LiBF}_4$	1
4G	GBL 68 vol %	VEC 2 vol %	EC 20 vol %	DEC 10 vol %	$\text{LiBF}_4$	1
5G	GBL 67 vol %	VEC 3 vol %	EC 20 vol %	DEC 10 vol %	$\text{LiBF}_4$	1

(EC= ethylene carbonate; VEC = vinyl ethylene carbonate; GBL = gamma-butyrolactone; DEC = diethyl carbonate; VC = vinylene carbonate)

10. As in Example 6, each of the eight batteries was charged at a constant current and constant voltage at an ambient temperature of 20°C, a fixed ceiling voltage of 4.2 V, and a maximum current of 1050 mA for 2.5 hours. The charged batteries were then discharged at a discharge current of 1500 mA at 20°C until the voltage reached 3.0 V. The amounts of gas generated in the batteries after the cycles were then measured.

### Results and Discussion

11. Table 2 shows the results obtained by measuring the amounts of gas generated after cycles for each of the eight batteries.

**Table 2**

<b>Battery</b>	<b>Component (B)</b>	<b>Amount of Gas Generated After Cycles (mL)</b>
<b>SERIES A BATTERIES</b>		
1	VEC and VC	2.4
2	VC	3.5
3	VEC	4.3
<b>SERIES B BATTERIES</b>		
1G	VEC and VC	1.9
2G	VC	3.0
3G	VEC	4.2
4G	VEC	4.0
5G	VEC	3.8

12. Considering first the three batteries in Series A which contain components (A), (B), and (C), it can be seen from Table 2 that battery 2, which contained only VC as component (B), generated 3.5 ml of gas. Battery 3 containing only VEC as component (B) produced 4.3 ml gas after cycles. However, when the cyclic carbonic acid ester (B) included *both* VC and VEC (battery 1), the amount of gas generated after the cycles was reduced to only 2.4 ml, a dramatic decrease of 31% relative to battery 2 containing only VC and a decrease of 44% relative to battery 3 containing only VEC. All of the batteries in Series A contain the same volume percentage of component (B) (3 vol %) and the same concentrations of GBL and EC.

13. Similar results were observed for the batteries of Series B, which also contain a component (D). As can be seen from comparing the data in Table 2, battery 2G, which included *only* VC as component (B), generated 3.0 mL of gas. Batteries 3G, 4G, and 5G containing *only* VEC as component (B) produced 3.8 to 4.2 ml gas after cycles. However, when the cyclic carbonic acid ester (B) included *both* VC and VEC (battery 1G), the amount of gas generated after the cycles was reduced to only 1.9 ml, a dramatic decrease of 37% relative to a battery containing only VC, and a decrease of 50-55% relative to batteries containing only VEC. Further, when the total amount of component (B) was a constant 3 vol % (as in batteries 1G and 5G), use of a combination of VEC and VC resulted in a dramatic 50% reduction in the amount of gas generated.

14. It can thus be concluded from Table 2 that the addition of VC and VEC to a solvent mixture containing a cyclic carboxylic acid ester ((A), here, GBL) and a cyclic carbonic acid ester ((C), here, EC) is much more effective at decreasing the amount of gas generated after cycles than when either VC or VEC is added alone to the same mixture of (A) and (C), regardless of the concentration of VEC. Similar results are observed when a linear carbonic acid ester ((D), here, DEC) is also included in the solvent mixture.

#### Conclusion

15. This Declaration demonstrates that by preparing non-aqueous electrolyte secondary batteries according to the invention, which contain VC and VEC in combination with a cyclic carboxylic acid ester (A), a cyclic carbonic acid ester (C) having no carbon-carbon unsaturated bond, and optionally a linear carbonic acid ester (D), favorable and unexpected results are obtained, particularly with regard to amount of gas generated after cycles. None of the prior art of record specifically teaches the use of VC and VEC in combination with components (A), (C), and optionally (D) in the non-aqueous solvent, nor makes obvious the results which have been observed by the present invention.

I declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further, that those statements were made with the knowledge that willful false statements the like so made are punishable by fine or imprisonment, or both, under Section 1003 of Title 18 of the United States Code; and that such willful statements may jeopardize the validity of the application or any patent issuing thereon.

Dated: 2005. June. 15

Atsushi Ueda  
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